6

RADIOACTIVE DECAY

The radioactive decays of naturally occurring minerals containing uranium and thorium are in large part responsible for the birth of the study of nuclear physics. These decays have half-lives that are of the order of the age of the Earth, suggesting that the materials are survivors of an early period in the creation of matter by aggregation of nucleons; the shorter-lived nuclei have long since decayed away, and we observe today the remaining long-lived decays. Were it not for the extremely long half-lives of ^{235}U and ^{238}U, we would today find no uranium in nature and would probably have no nuclear reactors or nuclear weapons.

In addition to this naturally occurring radioactivity, we also have the capability to produce radioactive nuclei in the laboratory through nuclear reactions. This was first done in 1934 by Irene Curie and Pierre Joliot, who used α particles from the natural radioactive decay of polonium to bombard aluminum, thereby producing the isotope ^{30}P, which they observed to decay through positron emission with a half-life of 2.5 min. In their words:

> Our latest experiments have shown a very striking fact: when an aluminum foil is irradiated on a polonium preparation, the emission of positrons does not cease immediately when the active preparation is removed. The foil remains radioactive and the emission of radiation decays exponentially as for an ordinary radioelement.

For this work on artificially produced radioactivity the Joliot-Curie team was awarded the 1935 Nobel Prize in Chemistry (following a family tradition—Irene's parents, Pierre and Marie Curie, shared with Becquerel the 1903 Nobel Prize in Physics for their work on the natural radioactivity of the element radium, and Marie Curie became the first person twice honored, when she was awarded the 1911 Nobel Prize in Chemistry).

In this chapter we explore the physical laws governing the production and decay of radioactive materials, which we take to mean those substances whose nuclei spontaneously emit radiations and thereby change the state of the nucleus.

6.1 THE RADIOACTIVE DECAY LAW

Three years following the 1896 discovery of radioactivity it was noted that the decay rate of a pure radioactive substance decreases with time according to an exponential law. It took several more years to realize that radioactivity represents changes in the individual atoms and not a change in the sample as a whole. It took another two years to realize that the decay is statistical in nature, that it is impossible to predict when any given atom will disintegrate, and that this hypothesis leads directly to the exponential law. This lack of predictability of the behavior of single particles does not bother most scientists today, but this early instance of it, before the development of quantum theory, was apparently difficult to accept. Much labor was required of these dedicated investigators to establish what now may seem like evident facts.

If N radioactive nuclei are present at time t and if no new nuclei are introduced into the sample, then the number dN decaying in a time dt is proportional to N, and so

$$\lambda = -\frac{(dN/dt)}{N} \tag{6.1}$$

in which λ is a constant called the *disintegration or decay constant*. The right side of Equation 6.1 is the probability per unit time for the decay of an atom. *That this probability is constant, regardless of the age of the atoms, is the basic assumption of the statistical theory of radioactive decay.* (Human lifetimes do not follow this law!)

Integrating Equation 6.1 leads to the *exponential law of radioactive decay*

$$N(t) = N_0 e^{-\lambda t} \tag{6.2}$$

where N_0, the constant of integration, gives the original number of nuclei present at $t = 0$. The *half-life* $t_{1/2}$ gives the time necessary for half of the nuclei to decay. Putting $N = N_0/2$ in Equation 6.2 gives

$$t_{1/2} = \frac{0.693}{\lambda} \tag{6.3}$$

It is also useful to consider the *mean lifetime* (sometimes called just the lifetime) τ, which is defined as the average time that a nucleus is likely to survive before it decays. The number that survive to time t is just $N(t)$, and the number that decay between t and $t + dt$ is $|dN/dt|\,dt$. The mean lifetime is then

$$\tau = \frac{\int_0^\infty t\,|dN/dt|\,dt}{\int_0^\infty |dN/dt|\,dt} \tag{6.4}$$

where the denominator gives the total number of decays. Evaluating the integrals gives

$$\tau = \frac{1}{\lambda} \tag{6.5}$$

Thus the mean lifetime is simply the inverse of the decay constant.

Equation 6.2 allows us to predict the number of undecayed nuclei of a given type remaining after a time t. Unfortunately, the law in that form is of limited usefulness because N is a very difficult quantity to measure. Instead of counting the number of undecayed nuclei in a sample, it is easier to count the number of decays (by observing the emitted radiations) that occur between the times t_1 and t_2. If we deduce a change ΔN in the number of nuclei between t and $t + \Delta t$, then

$$|\Delta N| = N(t) - N(t + \Delta t) = N_0 e^{-\lambda t}(1 - e^{-\lambda \Delta t}) \tag{6.6}$$

If the interval Δt during which we count is much smaller than λ^{-1} (and thus, in effect, $\Delta t \ll t_{1/2}$), we can ignore higher order terms in the expansion of the second exponential, and

$$|\Delta N| = \lambda N_0 e^{-\lambda t} \Delta t \tag{6.7}$$

Going over to the differential limit gives

$$\left|\frac{dN}{dt}\right| = \lambda N_0 e^{-\lambda t} \tag{6.8}$$

Defining the *activity* $\mathscr{A}$ to be the rate at which decays occur in the sample,

$$\mathscr{A}(t) \equiv \lambda N(t) = \mathscr{A}_0 e^{-\lambda t} \tag{6.9}$$

The initial activity at $t = 0$ is $\mathscr{A}_0 = \lambda N_0$.

Actually, we could have obtained Equation 6.8 by differentiating Equation 6.2 directly, but we choose this more circuitous path to emphasize an important but often overlooked point: *Measuring the number of counts ΔN in a time interval Δt gives the activity of the sample only if $\Delta t \ll t_{1/2}$.* The number of decays in the interval from t_1 to t_2 is

$$\Delta N = \int_{t_1}^{t_2 = t_1 + \Delta t} \mathscr{A}\, dt \tag{6.10}$$

which equals $\mathscr{A}\Delta t$ only if $\Delta t \ll t_{1/2}$. (Consider an extreme case—if $t_{1/2} = 1$ s, we observe the same number of counts in 1 min as we do in 1 h.) See Problem 1 at the end of this chapter for more on the relation between $\mathscr{A}$ and ΔN.

The activity of a radioactive sample is exactly the number of decays of the sample per unit time, and decays/s is a convenient unit of measure. Another unit for measuring activity is the *curie* (Ci), which originally indicated the activity of one gram of radium but is now defined simply as

$$1 \text{ Ci} = 3.7 \times 10^{10} \text{ decays/s}$$

Most common radioactive sources of strengths typically used in laboratories have activities in the range of microcuries to millicuries. The SI unit for activity is the becquerel (Bq), equal to one decay per second; however, the curie is so firmly in place as a unit of activity that the becquerel has not yet become the commonly used unit.

Note that the activity tells us only the number of disintegrations per second; it says nothing about the *kind* of radiations emitted or their *energies*. If we want to know about the effects of radiation on a biological system, the activity is not a useful quantity since different radiations may give different effects. In Section 6.8 we discuss some alternative units for measuring radiation that take into account their relative biological effects.

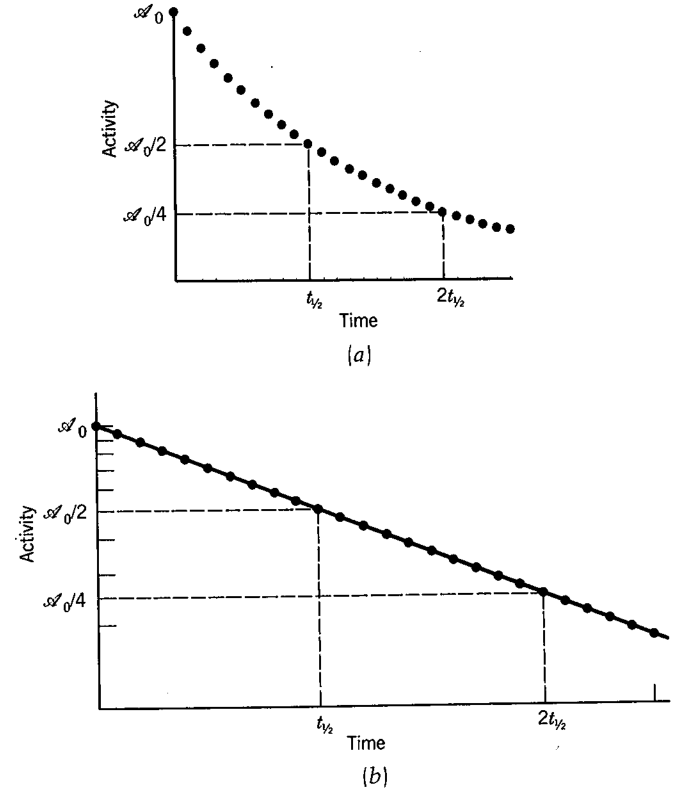

Figure 6.1 The exponential decay of activity. (*a*) Linear plot. (*b*) Semilog plot.

Equation 6.9 shows that the activity decays exponentially with time. We can thus measure the activity as a function of time by counting the number of decays in a sequence of short time intervals Δt. Plotting these data on a semilog graph (that is, $\ln \mathscr{A}$ vs t) should give a straight line of slope $-\lambda$. Figure 6.1 is an example of this kind of experiment, from which one can determine the half-life of a radioactive decay.

This method of measurement is useful only for half-lives that are neither too short nor too long. The half-life must be short enough that we can see the sample decaying—for half-lives far greater than a human lifetime, we would not be able to observe any substantial reduction in activity. For such cases, we can use Equation 6.1 directly, by measuring dN/dt (which is just the activity in this simple decay process) and by determining the number of atoms (such as by weighing a sample whose chemical composition is accurately known).

For half-lives that are very short (say, small compared with 1 s), observing the successive disintegration rates is also not useful, for the activity decays to nothing in the time that it would take to switch the counting apparatus on and off. For these cases we use a more precise technique, described in Chapter 7, that permits the routine measurement of half-lives down to nanoseconds (10^{-9} s) and even picoseconds (10^{-12} s).

It is important to keep in mind that the simple exponential law of radioactive decay applies only in a limited set of circumstances—a given initial quantity of a substance decays (by emitting radiation) to a stable end product. Under these circumstances, when radioactive nucleus 1 decays with decay constant λ_1 to stable nucleus 2, the number of nuclei present is

$$N_1 = N_0 e^{-\lambda_1 t} \tag{6.11a}$$

$$N_2 = N_0(1 - e^{-\lambda_1 t}) \tag{6.11b}$$

Note that the number of nuclei of type 2 starts out at 0 and approaches N_0 as $t \to \infty$ (all of type 1 eventually end as type 2) and also note that $N_1 + N_2 = N_0$ (the total number of nuclei is constant). If nuclei of type 2 are themselves radioactive, or if nuclei of type 1 are being produced (as a result of a nuclear reaction, for instance) then Equations 6.11 do not apply. We consider these cases in Sections 6.3 and 6.4.

Often it will happen that a given initial nucleus can decay in two or more different ways, ending with two different final nuclei. Let's call these two decay modes a and b. The rate of decay into mode a, $(dN/dt)_a$, is determined by the *partial decay constant* λ_a, and the rate of decay into mode b, $(dN/dt)_b$, by λ_b:

$$\begin{aligned} \lambda_a &= \frac{-(dN/dt)_a}{N} \\ \lambda_b &= \frac{-(dN/dt)_b}{N} \end{aligned} \tag{6.12}$$

The *total decay rate* $(dN/dt)_t$ is

$$-\left(\frac{dN}{dt}\right)_t = -\left(\frac{dN}{dt}\right)_a - \left(\frac{dN}{dt}\right)_b = N(\lambda_a + \lambda_b) = N\lambda_t \tag{6.13}$$

where $\lambda_t = \lambda_a + \lambda_b$ is the *total decay constant*. The nuclei therefore decay according to $N = N_0 e^{-\lambda_t t}$, and the activity $|dN/dt|$ decays with decay constant λ_t. *Whether we count the radiation leading to final states a or b, we observe only the total decay constant* λ_t; we never observe an exponential decay of the activity with constants λ_a or λ_b. The relative decay constants λ_a and λ_b determine the probability for the decay to proceed by mode a or b. Thus a fraction λ_a/λ_t of the nuclei decay by mode a and a fraction λ_b/λ_t decay by mode b, so that

$$\begin{aligned} N_1 &= N_0 e^{-\lambda_{1,t} t} \\ N_{2,a} &= (\lambda_a/\lambda_t) N_0 (1 - e^{-\lambda_{1,t} t}) \\ N_{2,b} &= (\lambda_b/\lambda_t) N_0 (1 - e^{-\lambda_{1,t} t}) \end{aligned} \tag{6.14}$$

The separate factors λ_a or λ_b never appear in any exponential term; we cannot "turn off" one decay mode to observe the exponential decay of the other.

Another special case is that of a sample with two or more radionuclei with genetically unrelated decay schemes. Consider a mixture of ^{64}Cu (12.7 h) and ^{61}Cu (3.4 h); such mixtures cannot be chemically separated of course. The activity of a particular mixture is plotted against time on semilog paper in Figure 6.2. At the right end of the curve we assume (because the curve is linear) that only one activity is present; the limiting slope shows a 12.7-h half-life. By (1) extending this limiting slope backward, (2) taking differences between the curve and this straight line at various abscissas, and (3) plotting these differences on the same scale, we get the dot-dashed straight line that represents the 3.4-h half-life. The intercepts of both straight lines on the vertical axis give the initial counting rates for each component. This method can be extended to mixtures with more than two components, if the half-lives are sufficiently different from one another.

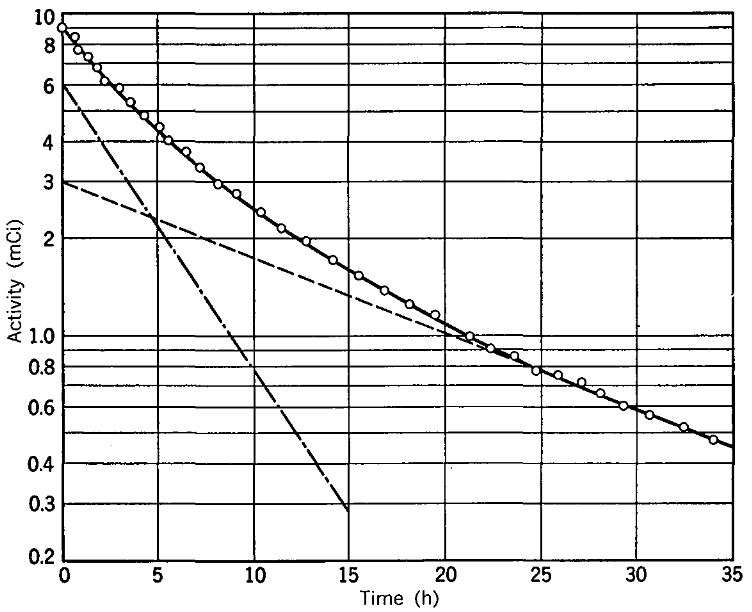

Figure 6.2 Decay curve for a sample containing a mixture of ^{64}Cu (12.7 h) and ^{61}Cu (3.4 h).

6.2 QUANTUM THEORY OF RADIATIVE DECAYS

The energy levels we obtain by solving the Schrödinger equation for various time-independent potentials share one property—they are *stationary states*. A quantum system that is originally in a particular stationary state will remain in that state for all times and will *not* make transitions to (i.e., decay to) other states. We can allow a quantum system to be found sometimes in one state and sometimes in another by making a mixture of two or more states, such as $\psi = c_1\psi_1 + c_2\psi_2$ which has the probability $|c_1|^2$ to be found in state 1 and $|c_2|^2$ to be found in state 2. For time-independent potentials, c_1 and c_2 are independent of time, which does not correspond with observations for decaying states, in which the probability to find one state decays with time. Moreover, on a philosophical level, we should be forced to abandon the notion of pure states with well-defined wave functions, making the interpretation of nuclear structure very difficult indeed.

We therefore adopt the following approach: The potential is assumed to be of the form $V + V'$, where V is the nuclear potential that gives the stationary states and V' is a very weak additional potential that can cause transitions between the states. For the moment neglecting V', we solve the Schrödinger equation for the potential V and obtain the static nuclear wave functions. We then use those wave functions to calculate the transition probability between the "stationary states" under the influence of V'. This transition probability is just the decay constant λ, which is given by Fermi's Golden Rule as discussed in Section 2.8:

$$\lambda = \frac{2\pi}{\hbar}|V'_{\mathrm{fi}}|^2\,\rho(E_{\mathrm{f}}) \tag{6.15}$$

where

$$V'_{\mathrm{fi}} = \int \psi_{\mathrm{f}}^{*} V' \psi_{\mathrm{i}}\, dv \tag{6.16}$$

Given the initial and final wave functions ψ_{i} and ψ_{f}, we can evaluate the "matrix element" of V' and thus calculate the transition probability (which can then be compared with its experimental value).

The transition probability is also influenced by the *density of final states* $\rho(E_{\mathrm{f}})$—within an energy interval dE_{f}, the number of final states accessible to the system is $dn_{\mathrm{f}} = \rho(E_{\mathrm{f}})\,dE_{\mathrm{f}}$. The transition probability will be large if there is a large number of final states accessible for the decay. There are two contributions to the density of final states because the final state after the decay includes two components—the final nuclear state and the emitted radiation. Let's consider in turn each of these two components, beginning with the nuclear state.

Solving the Schrödinger equation for the time-independent potential V gives us the stationary states of the nucleus, $\psi_{\mathrm{a}}(\boldsymbol{r})$. The time-dependent wave function $\Psi_{\mathrm{a}}(\boldsymbol{r}, t)$ for the state a is

$$\Psi_{\mathrm{a}}(\boldsymbol{r}, t) = \psi_{\mathrm{a}}(\boldsymbol{r})\, e^{-iE_{\mathrm{a}}t/\hbar} \tag{6.17}$$

where E_{a} is the energy of the state. The probability of finding the system in the state a is $|\Psi_{\mathrm{a}}(\boldsymbol{r}, t)|^2$, which is independent of time for a stationary state. To be consistent with the radioactive decay law, we would like the probability of finding our decaying system in the state a to decrease with time like $e^{-t/\tau_{\mathrm{a}}}$:

$$|\Psi_{\mathrm{a}}(t)|^2 = |\Psi_{\mathrm{a}}(t=0)|^2\, e^{-t/\tau_{\mathrm{a}}} \tag{6.18}$$

where $\tau_{\mathrm{a}} = 1/\lambda_{\mathrm{a}}$ is the mean lifetime of the state whose decay constant is λ_{a}. We should therefore have written Equation 6.17 as

$$\Psi_{\mathrm{a}}(\boldsymbol{r}, t) = \psi_{\mathrm{a}}(\boldsymbol{r})\, e^{-iE_{\mathrm{a}}t/\hbar}\, e^{-t/2\tau_{\mathrm{a}}} \tag{6.19}$$

The price we pay for including the *real* exponential term in Ψ_{a} is the loss of the ability to determine exactly the energy of the state—we no longer have a stationary state. (Recall the energy-time uncertainty relationship, Equation 2.2. If a state lives forever, $\Delta t \to \infty$ and we can determine its energy exactly, since $\Delta E = 0$. If a state lives on the average for a time τ, we cannot determine its energy except to within an uncertainty of $\Delta E \sim \hbar/\tau$.) We can make this discussion more rigorous by calculating the distribution of energy states (actually the Fourier transform of $e^{-t/2\tau_{\mathrm{a}}}$). The probability to observe the system in the energy interval between E and $E + dE$ in the vicinity of E_{a} is given by the square of this distribution:

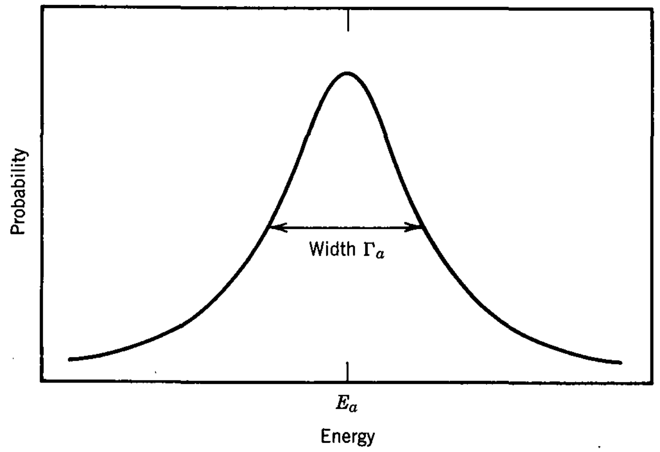

Figure 6.3 Probability to observe the energy of an unstable state of width Γ_a.

$$P(E)\,dE = \frac{dE}{(E - E_{\mathrm{a}})^2 + \Gamma_{\mathrm{a}}^2/4} \tag{6.20}$$

where $\Gamma_{\mathrm{a}} = \hbar/\tau_{\mathrm{a}}$ is the *width* of the state a. Figure 6.3 shows the function $P(E)$. If we measure the energy of this system, we may no longer find the value E_{a} (although the average of many measurements gives E_{a}). The width Γ_{a} is a measure of our inability to determine precisely the energy of the state (through no fault of our own—nature imposes the limit of uncertainty, not our measuring instruments; as indicated by Figure 6.3, a state with the "exact" energy E_{a} cannot be observed).

If nuclear states do not have exact energies, can we speak of transitions between distinct levels? We can, because *the widths of the low-lying nuclear levels are small compared with their energy spacing*. Nuclear states typically have lifetimes greater than 10^{-12} s, corresponding to $\Gamma < 10^{-10}$ MeV. The discrete low-lying nuclear states that are populated in ordinary decays (and many nuclear reactions, as well) have typical separations of the order of 10^{-3} MeV and larger. Thus if we were to measure the energy of a final nuclear state after a decay process (by measuring the energy of the emitted radiation, for example), it is very unlikely that the overlap of the energy distributions of two different final states a and b could cause confusion as to the final "stationary" state resulting from the decay (see Figure 6.4).

We therefore conclude that it *is* reasonable to speak of discrete pseudo-stationary states because their separation is far greater than their width, and we also conclude that such nuclear states do not contribute to the density of final states because there is only one nuclear state that can be reached in a given decay process.

It is thus only the radiation field that contributes to the density of states, and we must consider the properties of the emitted radiations in calculating $\rho(E_{\mathrm{f}})$. For the present, we will only make some general comments regarding $\rho(E_{\mathrm{f}})$. If

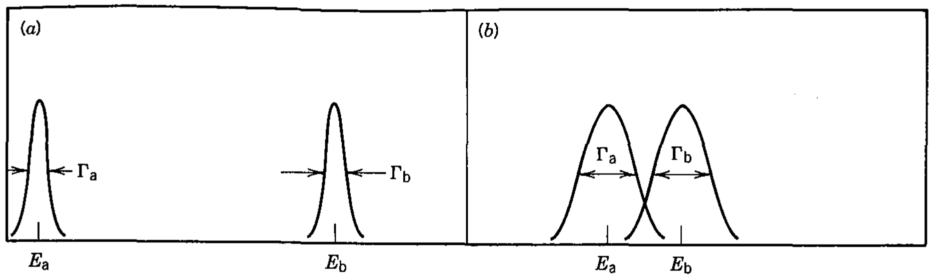

Figure 6.4 When the widths of unstable states are small compared with their separation, as in (*a*), the states are distinct and observable. In (*b*), the states a and b overlap and are strongly mixed; these states do not have distinctly observable wave functions.

we observe only the probability to form the nuclear state E_f, then we must consider all possible radiations of energy $E_i - E_f$. Specifically, the radiation can be emitted in any direction and in any state of polarization (if the radiation consists of a particle with spin, the spin may have any possible orientation), assuming of course that we do not observe the direction of the radiation or its polarization. It is this process of counting the number of accessible final states that gives the density of states, which we consider further when we discuss specific radiation types in Chapters 8–10.

In solving the differential equation (6.1) to obtain the radioactive decay law, we assumed the decay probability λ to be (1) small and (2) constant in time, which happen to be the same assumptions made in deriving Fermi's Golden Rule. If V' is independent of time, then λ calculated according to Equation 6.15 will also be independent of time. Under such a condition, the effect of V' on the stationary states a and b of V is

$$\psi_a \to \psi_a + \frac{V'_{ba}}{E_b - E_a}\psi_b$$

and the system formerly in the state a has a probability proportional to $|V'_{ba}|^2$ to be found in the state b. We observe this as a "decay" from a to b.

To apply Fermi's Golden Rule, the probability for decay must also be small, so that the amplitude of ψ_b in the above expression is small. It is this requirement that gives us a decay process. If the decay probability were large, then there would be enough radiation present to induce the reverse transition b → a through the process of resonant absorption. The system would then oscillate between the states a and b, in analogy with a classical system of two coupled oscillators.

The final connection between the effective decay probability for an ensemble of a large number of nuclei and the microscopic decay probability computed from the quantum mechanics of a single nucleus requires the assumption that each nucleus of the ensemble emits its radiation independently of all the others. We assume that the decay of a given nucleus is independent of the decay of its neighbors. This assumption then permits us to have confidence that the decay constant we measure in the laboratory can be compared with the result of our quantum mechanical calculation.

6.3 PRODUCTION AND DECAY OF RADIOACTIVITY

It quite frequently happens that a basic condition imposed in deriving the exponential law, that no new nuclei are introduced into the sample, is not valid. In solving Equation 6.1 we obtained a fixed number N_0 of nuclei present at $t = 0$. In many applications, however, we produce activity continuously, such as by a nuclear reaction. In this case, Equation 6.2 is no longer valid and we must consider in more detail the processes that occur in the production and decay of the activity.

Let's assume that we place a target of stable nuclei into a reactor or an accelerator such as a cyclotron. The nuclei of the target will capture a neutron or a charged particle, possibly leading to the production of a radioactive species. The rate R at which this occurs will depend on the number N_0 of target atoms present, on the flux or current I of incident particles, and on the reaction cross section σ (which measures the probability for one incident particle to react with one target nucleus). A typical flux of particles in a reactor or cyclotron might be of the order of $10^{14}/\mathrm{s} \cdot \mathrm{cm}^2$, and typical cross sections are at most of the order of barns ($10^{-24}\ \mathrm{cm}^2$). Thus the probability to convert a target particle from stable to radioactive is about $10^{-10}/\mathrm{s}$. Even if the reaction is allowed to continue for hours, the absolute number of converted target particles is small (say, less than 10^{-6} of the original number). *We can therefore, to a very good approximation, regard the number of target nuclei as constant*, and under this approximation the rate R is constant. (As we "burn up" target nuclei, N_0 will decrease by a small amount and the rate may therefore similarly decrease with time. Obviously N_0 must go to zero as $t \to \infty$, but for ordinary reaction times and typical cross sections we ignore this very small effect.) Thus

$$R = N_0 \sigma I \tag{6.21}$$

is taken to be a constant giving the rate at which the radioactive product nuclei are formed.

Let's denote by N_1 the number of radioactive nuclei that are formed as a result of the reaction. These nuclei decay with decay constant λ_1 to the stable nuclei denoted by N_2. Thus the number of nuclei N_1 present *increases* owing to the production at the rate R and *decreases* owing to the radioactive decay:

$$dN_1 = R\,dt - \lambda_1 N_1\,dt \tag{6.22}$$

and the solution to this equation is easily obtained

$$N_1(t) = \frac{R}{\lambda_1}(1 - e^{-\lambda_1 t}) \tag{6.23}$$

and

$$\mathscr{A}_1(t) = \lambda_1 N_1(t) = R(1 - e^{-\lambda_1 t}) \tag{6.24}$$

If the irradiation time is short compared with one half-life, then we can expand the exponential and keep only the term linear in t:

$$\mathscr{A}_1(\mathrm{t}) \cong R\lambda_1 t \qquad t \ll t_{1/2} \tag{6.25}$$

For small times, the activity thus increases at a constant rate. This corresponds to the linear (in time) accumulation of product nuclei, whose number is not yet seriously depleted by radioactive decays.

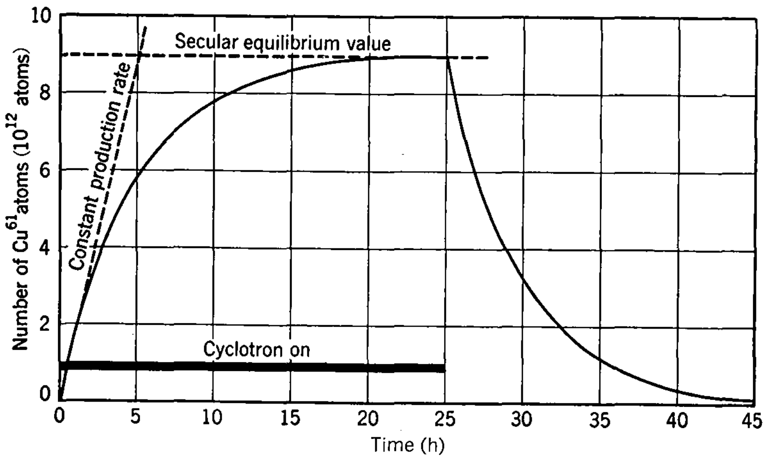

Figure 6.5 A plot of the number of radioactive ^{61}Cu atoms present in a Ni target at various times during and after bombardment with deuterons in a cyclotron.

For times long compared with the half-life the exponential approaches zero and the activity is approximately constant:

$$\mathscr{A}_1(t) \cong R \qquad t \gg t_{1/2} \tag{6.26}$$

In this case new activity is being formed at the same rate at which the older activity decays. This is an example of *secular equilibrium* which we discuss in more detail in the next section.

If we irradiate the sample for a time t_1 and then remove it from the accelerator or reactor, it will decay according to the simple exponential law, since no new activity is being formed. Figure 6.5 shows the activity resulting from the deuteron bombardment of ^{61}Ni to form ^{61}Cu ($t_{1/2} = 3.4$ h).

From Equation 6.24 we see that we produce 75% of the maximum possible activity by irradiating for two half-lives and 87.5% by irradiating for three half-lives. Further irradiation increases the activity by a steadily diminishing amount, so that we gain relatively little additional activity by irradiating for more than 2–3 half-lives. In fact, since the cost of using a reactor or accelerator is usually in direct proportion to the irradiation time, the best value (maximum activity per unit cost) is obtained by remaining close to the linear regime ($t \ll t_{1/2}$).

6.4 GROWTH OF DAUGHTER ACTIVITIES

Another common situation occurs when a radioactive decay results in a product nucleus that is also radioactive. It is thus possible to have series or chains of radioactive decays $1 \to 2 \to 3 \to 4 \ldots$, and it has become common to refer to the original nucleus (type 1) as the parent and the succeeding "generations" as daughter (type 2), granddaughter (type 3), and so on.

We assume that we begin with N_0 atoms of the parent at $t = 0$ and that no atoms of the decay products are originally present:

$$N_1(t=0) = N_0$$
$$N_2(t=0) = N_3(t=0) = \cdots = 0 \tag{6.27}$$

The various decay constants are represented by $\lambda_1, \lambda_2, \lambda_3, \ldots$. For the present calculation, we will assume that the granddaughter is the stable end-product of the decay. The number of parent nuclei decreases with time according to the usual form

$$dN_1 = -\lambda_1 N_1\, dt \tag{6.28}$$

The number of daughter nuclei increases as a result of decays of the parent and decreases as a result of its own decay:

$$dN_2 = \lambda_1 N_1\, dt - \lambda_2 N_2\, dt \tag{6.29}$$

The number of parent nuclei can be found directly from integrating Equation 6.28:

$$N_1(t) = N_0 e^{-\lambda_1 t} \tag{6.30}$$

To solve Equation 6.29, we try a solution of the form $N_2(t) = A e^{-\lambda_1 t} + B e^{-\lambda_2 t}$ and by substituting into Equation 6.29 and using the initial condition $N_2(0) = 0$ we find

$$N_2(t) = N_0 \frac{\lambda_1}{\lambda_2 - \lambda_1}\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) \tag{6.31}$$

$$\mathscr{A}_2(t) \equiv \lambda_2 N_2(t) = N_0 \frac{\lambda_2 \lambda_1}{\lambda_2 - \lambda_1}\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) \tag{6.32}$$

Note that Equation 6.31 reduces to Equation 6.11*b* if nuclei of type 2 are stable ($\lambda_2 \to 0$). We can also include the results of the previous section as a special case of Equation 6.31. Let's suppose that λ_1 is very small (but not quite zero), so that $N_1 \cong N_0 - N_0 \lambda_1 t$. In a nuclear reaction, the number of target nuclei decreases at the rate R according to $N_0 - Rt$, and thus identifying $N_0 \lambda_1$ with R and neglecting λ_1 in comparison with λ_2, Equation 6.31 reduces to Equation 6.24 for the activity of type 2.

$\lambda_1 \ll \lambda_2$

In this case the parent is so long-lived that it decays at an essentially constant rate; for all practical times $e^{-\lambda_1 t} \cong 1$ and

$$N_2(t) \cong N_0 \frac{\lambda_1}{\lambda_2}\left(1 - e^{-\lambda_2 t}\right) \tag{6.33}$$

which is of the same form as Equation 6.24. Thus the activity $\mathscr{A}_2$ approaches the limiting value $N_0 \lambda_1$ as was shown in Figure 6.5.

This is another example of *secular equilibrium*, where as t becomes large nuclei of type 2 are decaying at the same rate at which they are formed: $\lambda_2 N_2 = \lambda_1 N_1$. (Note that Equation 6.29 shows immediately that $dN_2/dt = 0$ in this case.) Figure 6.6 shows an example of approximate secular equilibrium.

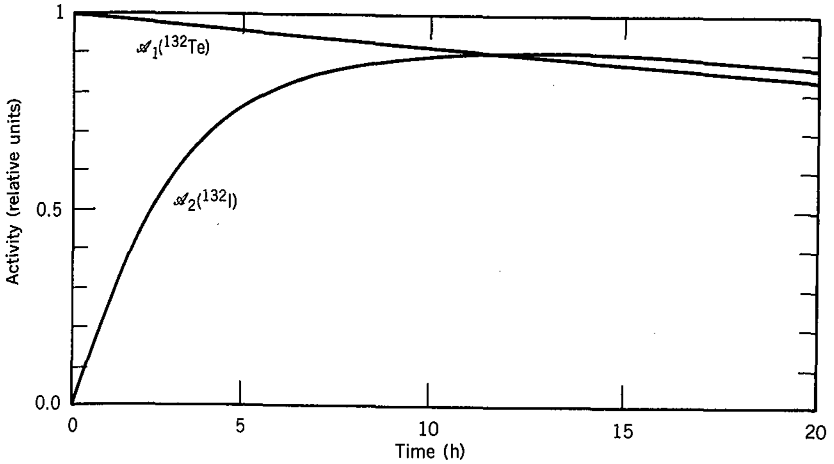

Figure 6.6 In the decay ^{132}Te (78 h) → ^{132}I (2.28 h) → ^{132}Xe, approximate secular equilibrium is reached at about 12 h.

$\lambda_1 < \lambda_2$

From Equations 6.30 and 6.31 we can calculate the ratio of the two activities:

$$\frac{\lambda_2 N_2}{\lambda_1 N_1} = \frac{\lambda_2}{\lambda_2 - \lambda_1}\left(1 - e^{-(\lambda_2 - \lambda_1)t}\right) \tag{6.34}$$

As t increases, the exponential term becomes smaller and the ratio $\mathscr{A}_2/\mathscr{A}_1$ approaches the limiting constant value $\lambda_2/(\lambda_2 - \lambda_1)$. The activities themselves are not constant, but the nuclei of type 2 decay (in effect) with the decay constant of type 1. This situation is known as *transient equilibrium* and is illustrated in Figure 6.7.

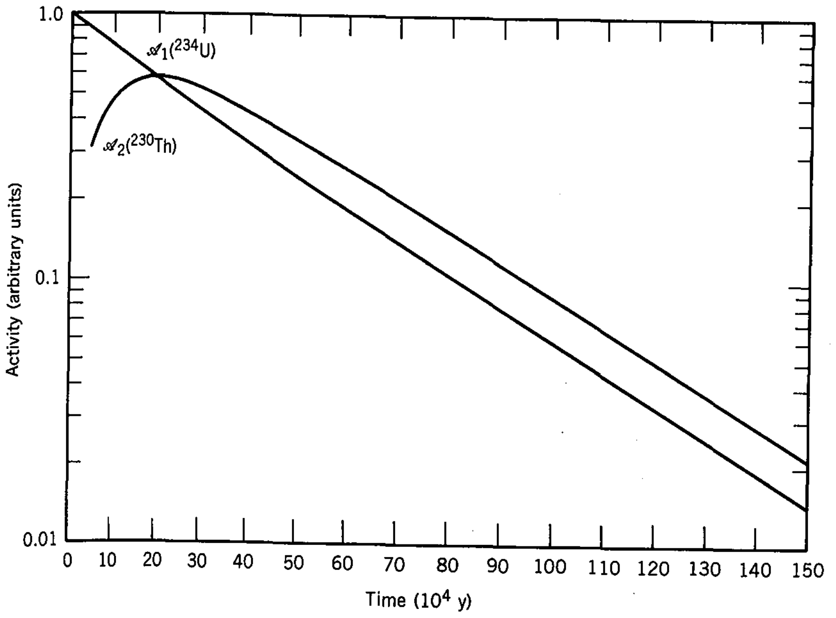

Figure 6.7 An example of equilibrium in the decays of ^{234}U (2.45×10^5 y) to ^{230}Th (8.0×10^4 y). The ratio $\mathscr{A}_2/\mathscr{A}_1$ approaches the constant value 1.48.

$\lambda_1 > \lambda_2$

In this case the parent decays quickly, and the daughter activity rises to a maximum and then decays with its characteristic decay constant. When this occurs the number of nuclei of type 1 is small and nearly insignificant. If t is so long that $e^{-\lambda_1 t}$ effectively vanishes, then Equation 6.31 becomes

$$N_2(t) \cong N_0 \frac{\lambda_1}{\lambda_1 - \lambda_2} e^{-\lambda_2 t} \tag{6.35}$$

which reveals that the type 2 nuclei decay approximately according to the exponential law.

Series of Decays

If we now assume that there are several succeeding generations of radioactive nuclei (that is, the granddaughter nuclei type 3 are themselves radioactive, as are types 4, 5, 6, ...), we can then easily generalize Equation 6.29 since each species is populated by the preceding one:

$$dN_i = \lambda_{i-1} N_{i-1}\, dt - \lambda_i N_i\, dt \tag{6.36}$$

A general solution, for the case of N_0 nuclei of type 1 and none of the other types initially present, is given by the *Bateman equations*, in which the activity of the nth member of the chain is given in terms of the decay constants of all preceding members:

$$\begin{aligned} \mathscr{A}_n &= N_0 \sum_{i=1}^{n} c_i e^{-\lambda_i t} \\ &= N_0\left(c_1 e^{-\lambda_1 t} + c_2 e^{-\lambda_2 t} + \cdots + c_n e^{-\lambda_n t}\right) \end{aligned} \tag{6.37}$$

where

$$\begin{aligned} c_m &= \frac{\prod_{i=1}^{n} \lambda_i}{\prod_{i=1}^{n}{}' (\lambda_i - \lambda_m)} \\ &= \frac{\lambda_1 \lambda_2 \lambda_3 \cdots \lambda_n}{(\lambda_1 - \lambda_m)(\lambda_2 - \lambda_m) \cdots (\lambda_n - \lambda_m)} \end{aligned} \tag{6.38}$$

where the prime on the lower product indicates we omit the term with $i = m$.

It is also possible to have secular equilibrium in this case, with $\lambda_1 N_1 = \lambda_2 N_2 = \cdots = \lambda_n N_n$.

6.5 TYPES OF DECAYS

The three primary decay types, to be discussed in greater detail in Chapters 8, 9, and 10, are α, β, and γ decays. In α- and β-decay processes, an unstable nucleus emits an α or a β particle as it tries to become a more stable nucleus (that is, to approach the most stable isobar for the resulting mass number). In γ-decay processes, an excited state decays toward the ground state without changing the nuclear species.

α Decay

In this process, a nucleus emits an α particle (which Rutherford and his co-workers showed to be a nucleus of helium, ${}^4_2\mathrm{He}_2$). The ${}^4\mathrm{He}$ nucleus is chosen as the agent for this process because it is such a tightly bound system, and thus the kinetic energy released in the decay is maximized. Such decays are favored, as we shall discuss in Chapter 8. The decay process is

$$ {}^A_Z\mathrm{X}_N \to {}^{A-4}_{Z-2}\mathrm{X}'_{N-2} + {}^4_2\mathrm{He}_2 $$

where X and X′ represent the chemical symbols of the initial and final nuclei. Notice that the number of protons and the number of neutrons must separately be conserved in the decay process. An example of an α-decay process is

$$ {}^{226}_{88}\mathrm{Ra}_{138} \to {}^{222}_{86}\mathrm{Rn}_{136} + \alpha $$

in which the half-life is 1600 years and the α particle appears with a kinetic energy of about 4.8 MeV.

β Decay

Here the nucleus can correct a proton or a neutron excess by directly converting a proton into a neutron or a neutron into a proton. This process can occur in three possible ways, each of which must involve another charged particle to conserve electric charge (the charged particle, originally called a β particle, was later shown to be identical with ordinary electrons).

$$ \mathrm{n} \to \mathrm{p} + \mathrm{e}^- \qquad \beta^- \text{ decay} $$

$$ \mathrm{p} \to \mathrm{n} + \mathrm{e}^+ \qquad \beta^+ \text{ decay} $$

$$ \mathrm{p} + \mathrm{e}^- \to \mathrm{n} \qquad \text{electron capture } (\varepsilon) $$

The first process is known as negative β decay or negatron decay and involves the creation and emission of an ordinary electron. The second process is positive β decay or positron decay, in which a positively charged electron is emitted. In the third process, an atomic electron that strays too close to the nucleus is swallowed, allowing the conversion of a proton to a neutron.

In all three processes, yet another particle called a *neutrino* is also emitted, but since the neutrino has no electric charge, its inclusion in the decay process does not affect the identity of the other final particles.

Note that in positive and negative β decay, a particle is created (out of the decay energy, according to $m = E/c^2$). The electron or positron did not exist inside the nucleus before the decay. (Contrast the case of α decay, in which the emitted nucleons were inside the nucleus before the decay.)

Some representative β-decay processes are

$$ {}^{131}_{53}\mathrm{I}_{78} \xrightarrow[\beta^-]{} {}^{131}_{54}\mathrm{Xe}_{77} \qquad t_{1/2} = 8.0 \text{ d} $$

$$ {}^{25}_{13}\mathrm{Al}_{12} \xrightarrow[\beta^+]{} {}^{25}_{12}\mathrm{Mg}_{13} \qquad t_{1/2} = 7.2 \text{ s} $$

$$ {}^{54}_{25}\mathrm{Mn}_{29} \xrightarrow[\varepsilon]{} {}^{54}_{24}\mathrm{Cr}_{30} \qquad t_{1/2} = 312 \text{ d} $$

In these processes, Z and N each change by one unit, but the total mass number $Z + N$ remains constant.

γ Decay

Radioactive γ emission is analogous to the emission of atomic radiations such as optical or X-ray transitions. An excited state decays to a lower excited state or possibly the ground state by the emission of a photon of γ radiation of energy equal to the difference in energy between the nuclear states (less a usually negligible correction for the "recoil" energy of the emitting nucleus). Gamma emission is observed in all nuclei that have excited bound states ($A > 5$), and usually follows α and β decays since those decays will often lead to excited states in the daughter nucleus.

The half-lives for γ emission are usually quite short, generally less than 10^{-9} s, but occasionally we find half-lives for γ emission that are significantly longer, even hours or days. These transitions are known as *isomeric transitions* and the long-lived excited states are called *isomeric states* or *isomers* (or sometimes *metastable* states). There is no clear criterion for classifying a state as isomeric or not; the distinction was originally taken to be whether or not the half-life was directly measurable, but today we can measure half-lives well below 10^{-9} s. Clearly a state with $t_{1/2} = 10^{-6}$ s is an isomer and one with $t_{1/2} = 10^{-12}$ s is not, but in between the boundary is rather fuzzy. We usually indicate metastable states with a superscript m, thus: ${}^{110}\mathrm{Ag}^{\mathrm{m}}$ or ${}^{110\mathrm{m}}\mathrm{Ag}$.

A process that often competes with γ emission is *internal conversion*, in which the nucleus de-excites by transferring its energy directly to an *atomic* electron, which then appears in the laboratory as a free electron. (This is very different from β decay in that no change of Z or N occurs, although the atom becomes ionized in the process.)

Spontaneous Fission

We usually think of fission as occurring under very unnatural and artificial conditions, such as in a nuclear reactor. There are, however, some nuclei that fission spontaneously, as a form of radioactive decay. This process is similar to the neutron-induced fission that occurs in reactors, with the exception that no previous neutron capture is needed to initiate the fission. In the process, a heavy nucleus with an excess of neutrons splits roughly in half into two lighter nuclei; the final nuclei are not rigidly determined, as they are in α or β decay, but are statistically distributed over the entire range of medium-weight nuclei. Examples of spontaneously fissioning nuclei are ^{256}Fm ($t_{1/2} = 2.6$ h) and ^{254}Cf ($t_{1/2} = 60.5$ days).

Nucleon Emission

As we move further and further from the "valley" of stable nuclei, the energy differences between neighboring isobars increases (recall the mass parabolas of constant A of Figure 3.18). Eventually the difference exceeds the nucleon binding energy (about 8 MeV, on the average) and it becomes possible to have radioactive decay by nucleon emission. This type of decay occurs most frequently in fission products, which have a very large neutron excess, and it is responsible for the "delayed" neutrons (that is, delayed by the half-life of the decay) that are used to control nuclear reactors. For example, ^{138}I β decays with a half-life of 6.5 s to ^{138}Xe. Most of the β decays populate low excited states in ^{138}Xe, but about 5% of the ^{138}I decays populate states in ^{138}Xe at about 6.5 MeV; these states decay by direct neutron emission to ^{137}Xe. Similarly, 0.7% of the ^{73}Kr β^+ decays ($t_{1/2} =$ 27 s) go to states in ^{73}Br at about 5 MeV; these states decay by proton emission to states in ^{72}Se.

Branching Ratios and Partial Half-lives

Figure 6.8 summarizes a variety of different decay processes, and Figure 6.9 shows a small section of the chart of stable and radioactive nuclei (Figure 1.1) with several decay processes indicated. Some nuclei may decay only through a single process, but more often decay schemes are very complicated, involving the emission of α's, β's, and γ's in competing modes. We specify the relative intensities of the competing modes by their *branching ratios*. Thus ^{226}Ra α decays to the ground state of ^{222}Rn with a branching ratio of 94% and to the first excited state with a branching ratio of 6%. Often different decay modes can compete: ^{226}Ac decays by α emission (0.006%), β^- emission (83%), and ε (17%); ^{132}Cs decays by β^- emission (2%) and by β^+ and ε (98%); the metastable state ^{95m}Nb decays by β^- emission (2.5%) or by an isomeric transition (97.5%). The isomeric transition itself includes a 27% branch by γ emission and a 73% branch by internal conversion.

Frequently, we specify the branching ratio by giving the partial decay constant or partial half-life. For example, we consider the decay of ^{226}Ac ($t_{1/2} = 29$ h). The total decay constant is

$$\lambda_t = \frac{0.693}{t_{1/2}} = 0.024\ \mathrm{h}^{-1} = 6.6 \times 10^{-6}\ \mathrm{s}^{-1}$$

The partial decay constants are

$$\lambda_\beta = 0.83\lambda_t = 5.5 \times 10^{-6}\ \mathrm{s}^{-1}$$

$$\lambda_\varepsilon = 0.17\lambda_t = 1.1 \times 10^{-6}\ \mathrm{s}^{-1}$$

$$\lambda_\alpha = 6 \times 10^{-5}\lambda_t = 4 \times 10^{-10}\ \mathrm{s}^{-1}$$

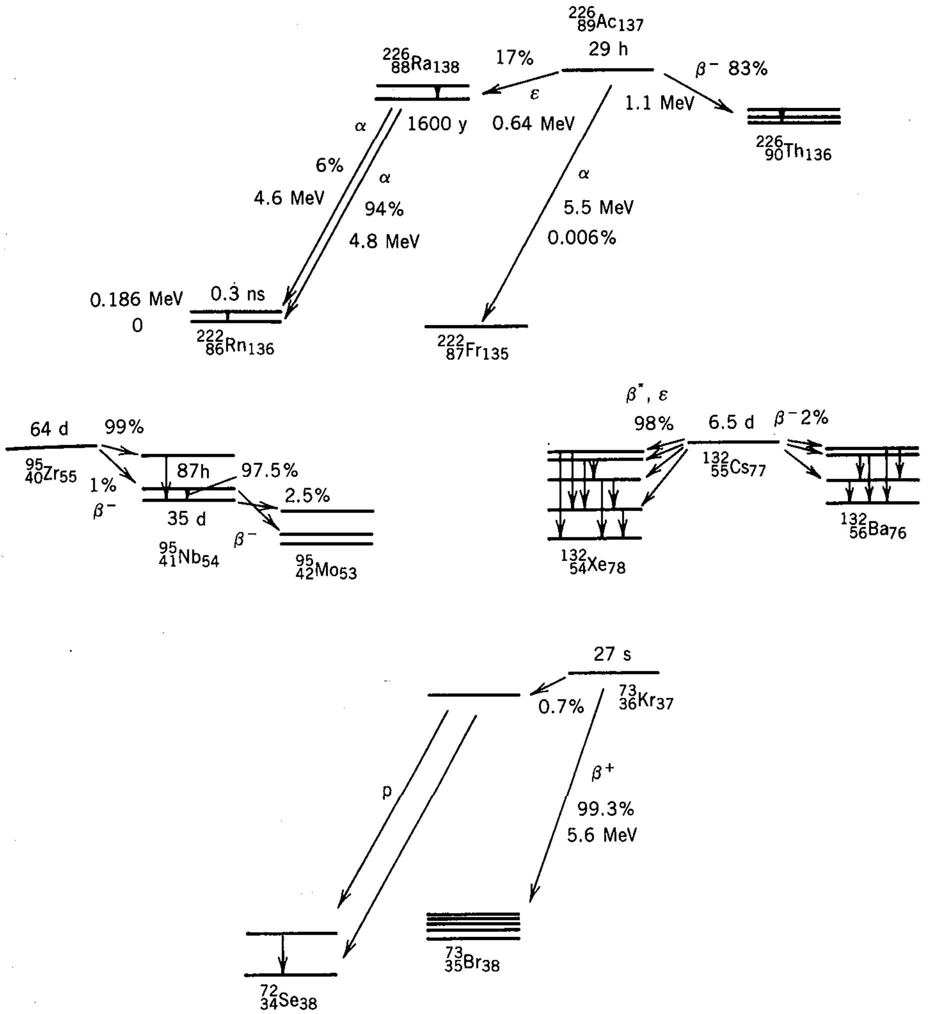

Figure 6.8 A variety of different decay processes.

and the partial half-lives are

$$t_{1/2,\beta} = \frac{0.693}{\lambda_\beta} = 1.3 \times 10^5\ \mathrm{s} = 35\ \mathrm{h}$$

$$t_{1/2,\varepsilon} = \frac{0.693}{\lambda_\varepsilon} = 6.1 \times 10^5\ \mathrm{s} = 170\ \mathrm{h}$$

$$t_{1/2,\alpha} = \frac{0.693}{\lambda_\alpha} = 1.7 \times 10^9\ \mathrm{s} = 55\ \mathrm{y}$$

The partial half-life is merely a convenient way to represent branching ratios; a glance at the above figures shows that α emission is far less probable than β emission for ^{226}Ac. *However, the activity would be observed to decay only with the total half-life.* Even if we were to observe the decay of ^{226}Ac by its α emission, the

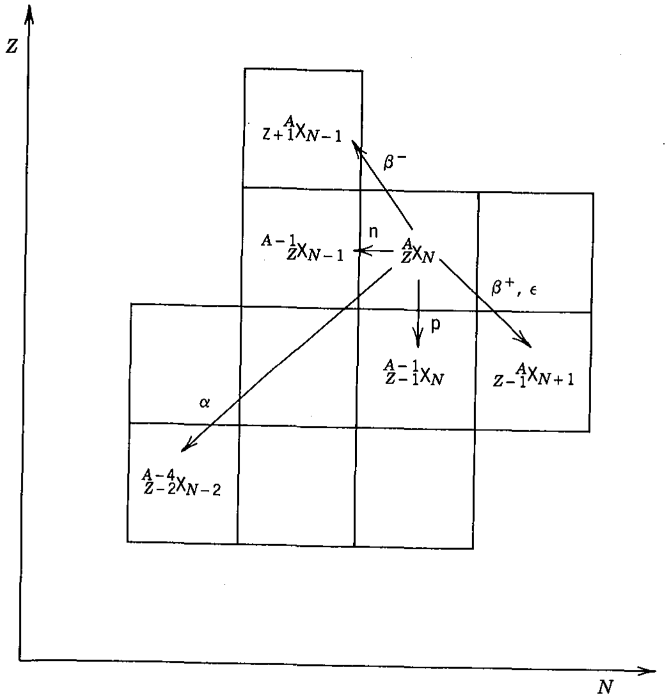

Figure 6.9 The initial nucleus $^{A}_{Z}X_N$ can reach different final nuclei through a variety of possible decay processes.

activity would decay with time according to a half-life of 29 h. (Imagine if this were not so, and two observers were studying the decay of ^{226}Ac, one by observing the β's and the other by observing the α's. Since the radioactive decay law gives the number of undecayed nuclei, the β observer would conclude that half of the original ^{226}Ac nuclei remained after 35 h, while the α observer would have to wait 55 years similarly to observe half of the nuclei undecayed! In reality, half of the nuclei decay every 29 h, no matter what method we use to observe those decays.)

6.6 NATURAL RADIOACTIVITY

The Earth and the other planets of our solar system formed about 4.5×10^9 y ago out of material rich in iron, carbon, oxygen, silicon, and other medium and heavy elements. These elements in turn were created from the hydrogen and helium that resulted from the Big Bang some 15×10^9 y ago. During the 10×10^9 y from the Big Bang until the condensation of the solar system, the hydrogen and helium were "cooked" into heavier elements in stellar interiors, novas, and supernovas; we are made of the recycled debris of these long dead stars. Most of the elements thus formed were radioactive, but have since decayed to stable nuclei. A few of the radioactive elements have half-lives that are long compared with the age of the Earth, and so we can still observe their radioactivity. This radioactivity forms the major portion of our natural radioactive environment, and is also probably responsible for the inner heating of the terrestrial planets.

Table 6.1 Some Characteristics of the Disintegration Series of the Heavy Elements

			Longest-Lived Member	
Name of Series	Type[a]	Final Nucleus (Stable)	Nucleus	Half-Life, (y)
Thorium	$4n$	^{208}Pb	^{232}Th	1.41×10^{10}
Neptunium	$4n + 1$	^{209}Bi	^{237}Np	2.14×10^6
Uranium	$4n + 2$	^{206}Pb	^{238}U	4.47×10^9
Actinium	$4n + 3$	^{207}Pb	^{235}U	7.04×10^8

[a] n is an integer.

Although there are long-lived natural radioactive elements of other varieties, most of those observed today originate with the very heavy elements, which have no stable isotopes at all. These nuclides decay by α and β emission, decreasing Z and A until a lighter, stable nucleus is finally reached. Alpha decay changes A by four units and β decay does not change A at all, and so therefore we have four independent decay chains with mass numbers $4n$, $4n + 1$, $4n + 2$, and $4n + 3$, where n is an integer. The decay processes will tend to concentrate the nuclei in the longest-lived member of the chain, and if the lifetime of that nuclide is at least of the order of the age of the Earth, we will observe that activity today. The four series are listed in Table 6.1. Notice that the longest-lived member of the neptunium series has far too short a half-life to have survived since the formation of the Earth; this series is not observed in natural material.

Consider, for example, the thorium series illustrated in Figure 6.10. Let us assume that we had created, in a short period of time, a variety of plutonium (Pu) isotopes. The isotopes ^{232}Pu and ^{236}Pu decay rapidly to 72-y ^{232}U and other species of much shorter half-lives. Thus in a time long compared with 72 y (say, 10^3 y), all traces of these isotopes have vanished, leaving only the stable end product ^{208}Pb. The isotopes ^{240}Pu and ^{244}Pu decay much more slowly, the former comparatively quickly and the latter very slowly to ^{236}U, which in turn decays to the longest-lived member of the series, ^{232}Th. In a time greater than 81×10^6 y but less than 14×10^9 y, the original ^{240}Pu and ^{244}Pu (and the intermediate ^{236}U) will all have decayed to ^{232}Th, the decay of which we still observe today.

These radioactive isotopes are present in material all around us, especially in rocks and minerals that condensed with the Earth 4.5×10^9 y ago. (In fact, their decays provide a reliable technique for determining the time since the condensation of the rocks and thus the age of the Earth; see Section 6.7 and Chapter 19 for discussions of these techniques.) In general the radioactive elements are tightly bound to the minerals and are not hazardous to our health, but all of the natural radioactive series involve the emission of a gaseous radioactive element, radon. This element, if formed deep within rocks, normally has little chance to migrate to the surface and into the air before it decays. However, when rocks are fractured, the radon gas can escape (in fact the presence of radon gas has in recent years been observed as a precursor of earthquakes). There is also the possibility of escape of radon from the surface of minerals, and particularly those that are used in the construction of buildings. Inhalation of this radioactive gas

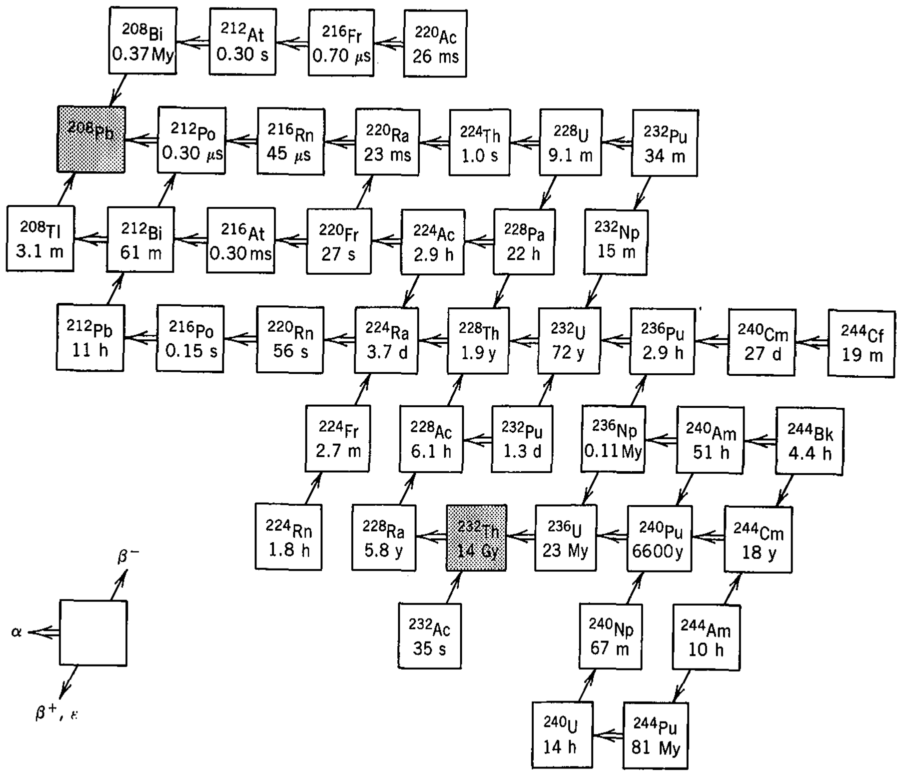

Figure 6.10 The thorium series of naturally occurring radioactive decays. Some half-lives are indicated in My (10^6 y) and Gy (10^9 y). The shaded members are the longest-lived radioactive nuclide in the series (Th, after which the series is named) and the stable end product.

Table 6.2 Some Natural Radioactive Isotopes

Isotope	$t_{1/2}$ (y)
^{40}K	1.28×10^9
^{87}Rb	4.8×10^{10}
^{113}Cd	9×10^{15}
^{115}In	4.4×10^{14}
^{138}La	1.3×10^{11}
^{176}Lu	3.6×10^{10}
^{187}Re	5×10^{10}

could possibly be responsible for many lung cancers, and there is a current suspicion that smoking may accelerate this process by causing the accumulation of these radioactive products in the lungs. It is perhaps ironic that the recent trends toward well insulated and tightly sealed buildings to conserve energy may be responsible for an increased concentration of radon gas, and as of this writing there is active research on the problem, including measurement of radon gas accumulation in buildings.

The heavy element series are not the only sources of naturally occurring radioactive isotopes of half-lives long enough to be present in terrestrial matter. Table 6.2 gives a partial list of others, some of which can also be used for radioactive dating.

There are also other natural sources of radioactivity of relatively short half-lives, which are not remnants of the production of elements before the Earth formed, but instead are being formed continuously today. These elements include ^{3}H and ^{14}C, which are formed in the upper atmosphere when cosmic rays (high-energy protons) strike atoms of the atmosphere and cause nuclear reactions. The isotope ^{14}C has had important applications in radioactive dating.

6.7 RADIOACTIVE DATING

Although we cannot predict with certainty when an individual nucleus will decay, we can be very certain how long it will take for half of a large number of nuclei to decay. These two statements may seem inconsistent; their connection has to do with the statistical inferences that we can make by studying random processes. If we have a room containing a single gas molecule, we cannot predict with certainty whether it will be found in the left half of the room or the right half. If however we have a room containing a large number N of molecules ($N \sim 10^{24}$), then we expect to find on the average $N/2$ molecules in each half. Furthermore, the fluctuations of the number in each half about the value $N/2$ are of the order of $\sqrt{N}$; thus the deviation of the fraction in each half from the value 0.5 is about $\sqrt{N}/N \simeq 10^{-12}$. The fraction in each half is thus $0.500000000000 \pm 0.000000000001$. This extreme (and unreasonable) precision comes about because N is large and thus the fractional error $N^{-1/2}$ is small.

A similar situation occurs for radioactive decay. (The laws of counting statistics are discussed in detail in Chapter 7.) If we had at $t = 0$ a collection of a large number N_0 of radioactive nuclei, then after a time equal to one half-life, we should find that the remaining fraction is $\frac{1}{2} \pm N_0^{-1/2}$. Thus despite the apparently random nature of the decay process, the decay of radioactive nuclei gives us a very accurate and entirely reliable clock for recording the passage of time. That is, if we know the decay constant λ, the exponential decrease in activity of a sample can be used to measure time.

The difficulty in using this process occurs when we try to apply it to decays that occur over geological times ($\sim 10^9$ y) because in this case we do not measure the activity as a function of time. Instead, we use the relative number of parent and daughter nuclei observed at time t_1 (now) compared with the relative number at time t_0 (when the "clock" started ticking, usually when the material such as a rock or mineral condensed, trapping the parent nuclei in their present sites). In principle this process is rather simple. Given the decay of parent isotope P to daughter isotope D, we merely count (by chemical means, for instance) the present numbers of P and D atoms, $N_P(t_1)$ and $N_D(t_1)$:

$$N_D(t_1) + N_P(t_1) = N_P(t_0) \tag{6.39}$$

$$N_P(t_1) = N_P(t_0)\, e^{-\lambda(t_1 - t_0)} \tag{6.40}$$

$$\Delta t \equiv t_1 - t_0 = \frac{1}{\lambda} \ln \frac{N_P(t_0)}{N_P(t_1)}$$

$$\Delta t = \frac{1}{\lambda} \ln \left(1 + \frac{N_D(t_1)}{N_P(t_1)} \right) \tag{6.41}$$

Given the decay constant (which we can measure in the laboratory) and the present ratio of daughter to parent nuclei, the age of the sample is directly found, with a precision determined by our knowledge of λ and by the counting statistics for N_P and N_D.

Equations 6.39 and 6.40 contain assumptions that must be carefully tested before we can apply Equation 6.41 to determine the age of a sample. Equation 6.39 assumes that $N_D(t_0) = 0$—no daughter atoms are present at t_0—and also that the total number of atoms remains constant—no parent or daughter atoms escape from the mineral or solid in which they were originally contained. As we discuss below, we can modify the derivation of Δt to account for the daughter atoms present at t_0 (even though when we analyze the sample today at time t_1, we cannot tell which daughter atoms were originally present and which resulted from decays during Δt). Equation 6.40 assumes that the variation in N_P comes only from the decay—no new parent atoms are introduced (by a previous decay or by nuclear reactions induced by cosmic rays, for example).

Let's relax the assumption of Equation 6.39 and permit daughter nuclei to be present at $t = t_0$. These daughter nuclei can be formed from the decay of parent nuclei at times before t_0 or from the process that formed the original parent nuclei (a supernova explosion, for example); the means of formation of these original daughter nuclei is of no importance for our calculation. We therefore take

$$N_D(t_1) + N_P(t_1) = N_D(t_0) + N_P(t_0) \tag{6.42}$$

Because we have introduced another unknown, $N_D(t_0)$, we can no longer solve directly for the age Δt. If, however, there is also present in the sample a different isotope of the daughter, D′, which is neither radioactive nor formed from the decay of a long-lived parent, we can again find the age of the sample. The population of this stable isotope is represented by $N_{D'}$, and if D′ is stable then $N_{D'}(t_1) = N_{D'}(t_0)$, in which case

$$\frac{N_D(t_1) + N_P(t_1)}{N_{D'}(t_1)} = \frac{N_D(t_0) + N_P(t_0)}{N_{D'}(t_0)} \tag{6.43}$$

which can be written as

$$\frac{N_D(t_1)}{N_{D'}(t_1)} = \frac{N_P(t_1)}{N_{D'}(t_1)}\left[e^{\lambda(t_1 - t_0)} - 1\right] + \frac{N_D(t_0)}{N_{D'}(t_0)} \tag{6.44}$$

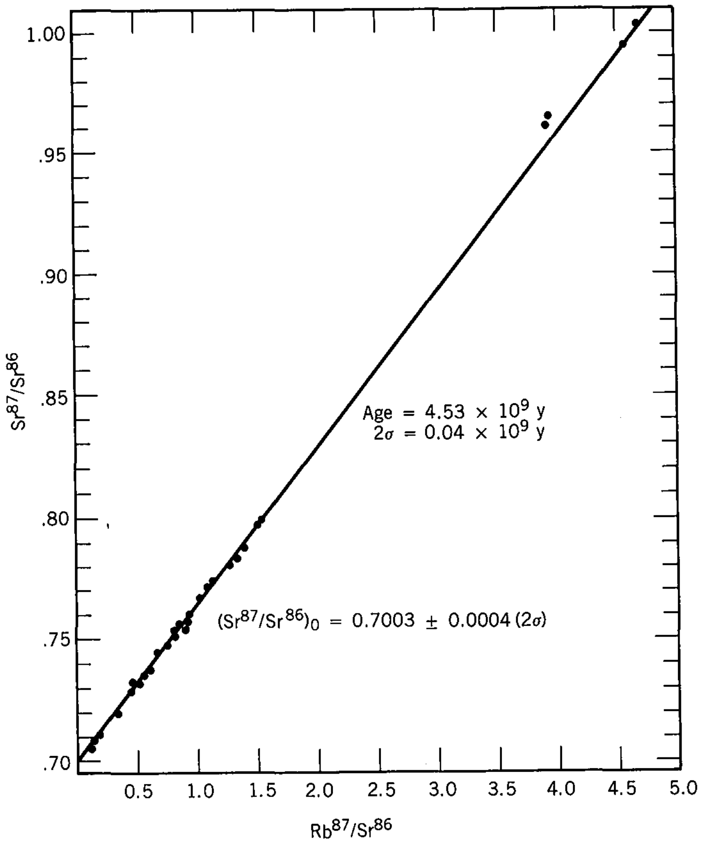

Figure 6.11 The Rb-Sr dating method, allowing for the presence of some initial ^{87}Sr. The linear behavior is consistent with Equation 6.44. From G. W. Wetherill, *Ann. Rev. Nucl. Sci.* **25**, 283 (1975).

The ratios $N_D(t_1)/N_{D'}(t_1)$ and $N_P(t_1)/N_{D'}(t_1)$ can be measured in the laboratory, but that still leaves two unknowns in Equation 6.44: the age Δt and the initial isotopic ratio $N_D(t_0)/N_{D'}(t_0)$. Minerals that crystallize from a common origin should show identical ages and identical isotopic ratios $N_D(t_0)/N_{D'}(t_0)$, even though the original $N_P(t_0)$ may be very different (from differing chemical compositions, for example). If these hypotheses are correct, we expect to observe today minerals with various ratios $N_D(t_1)/N_{D'}(t_1)$ and $N_P(t_1)/N_{D'}(t_1)$ corresponding to common values of Δt and $N_D(t_0)/N_{D'}(t_0)$. We can test these assumptions by plotting $y = N_D(t_1)/N_{D'}(t_1)$ against $x = N_P(t_1)/N_{D'}(t_1)$ for a variety of minerals. Equation 6.44 is of the form $y = mx + b$, a straight line with slope $m = e^{\lambda(t_1 - t_0)} - 1$ and intercept $b = N_D(t_0)/N_{D'}(t_0)$. Figure 6.11 is an example of such a procedure for the decay $^{87}\text{Rb} \rightarrow {}^{87}\text{Sr}$ ($t_{1/2} = 4.8 \times 10^{10}$ y), in which the comparison is done with stable ^{86}Sr. Even though the present ratio of ^{87}Rb to ^{86}Sr varies by more than an order of magnitude, the data indicate a common age of the Earth, $\Delta t = 4.5 \times 10^9$ y. The good linear fit is especially important, for it justifies our assumptions of no loss of parent or daughter nuclei.

Other similar methods for dating minerals from the Earth, Moon, and meteorites give a common age of 4.5×10^9 y. These methods include the decay of ^{40}K to ^{40}Ar, the decay of ^{235}U and ^{238}U to ^{207}Pb and ^{206}Pb, and the spontaneous fission of ^{238}U and ^{244}Pu, which are analyzed either by chemical separation of the fission products or by microscopic observation of the tracks left in the minerals by the fission fragments.

For dating more recent samples of organic matter, the ^{14}C dating method is used. The CO_2 that is absorbed by organic matter consists almost entirely of stable ^{12}C (98.89%), with a small mixture of stable ^{13}C (1.11%). Radioactive ^{14}C is being continuously formed in the upper atmosphere as a result of cosmic-ray bombardment of atmospheric nitrogen, and thus all living matter is slightly radioactive owing to its ^{14}C content. Because the production rate of ^{14}C by cosmic rays has been relatively constant for thousands of years, living organic material reaches equilibrium of its carbon with atmospheric carbon, with about 1 atom of ^{14}C for every 10^{12} atoms of ^{12}C. The half-life of ^{14}C is 5730 y, and thus each gram of carbon shows an activity of about 15 decays per minute. When an organism dies, it goes out of equilibrium with atmospheric carbon; it stops acquiring new ^{14}C and its previous content of ^{14}C decreases according to the radioactive decay law. We can therefore determine the age of samples by measuring the *specific activity* (activity per gram) of their carbon content. This method applies as long as we have enough ^{14}C intensity to determine the activity; from matter that has decayed for 10 or more half-lives, the decay is so weak that the ^{14}C method cannot be used. Recent techniques using accelerators as mass spectrometers have the potential to exceed this limit by counting ^{14}C atoms directly; these techniques are discussed in Chapter 20.

The major assumption of this method is the relatively constant production of ^{14}C by cosmic rays over the last 50,000 y or so. We can test this assumption by comparing the ages determined by radiocarbon dating with ages known or determined by independent means (historical records or tree-ring counting, for example). These comparisons show very good agreement and support the assumption of a relatively uniform flux of cosmic rays.

In later millennia, the radiocarbon method may no longer be applicable. During the last 100 years, the burning of fossil fuels has upset the atmospheric balance by diluting the atmosphere with stable carbon (the hydrocarbons of fossil fuels are old enough for all of their ^{14}C to have decayed away). During the 1950s and 1960s, atmospheric testing of nuclear weapons has placed additional ^{14}C in the atmosphere, perhaps doubling the concentration over the equilibrium value from cosmic-ray production alone.

6.8 UNITS FOR MEASURING RADIATION

The activity of a radioactive sample (in curies or in decays per second) does not depend on the type of radiation or on its energy. Thus the activity may be a useful means to compare two different sources of the same decaying isotope (10 mCi of ^{60}Co is stronger than 1 mCi of ^{60}Co), but how can we compare different decays? For instance, how does a 10-mCi source of ^{60}Co compare in strength with 10 mCi of ^{14}C, or how does a 10-μCi γ emitter compare in strength with a 10-mCi α emitter? And just what exactly do we mean by the "strength" of a source of radiation?

One common property of nuclear radiations is their ability to *ionize* (knock electrons from) atoms with which they interact. (For this reason, nuclear radiation is often called *ionizing radiation*.) We begin by considering the passage of X-ray and γ-ray photons through air. The photons interact many times with atoms in the air through a variety of processes (Compton scattering, photoelectric effect, electron-positron pair production), each of which creates a free electron, often of reasonably high energy. These secondary electrons can themselves produce ionization (and additional electrons). The total electric charge Q on the ions produced in a given mass m of air is called the *exposure* X, and we may take γ-ray sources as being of the same strength if they result in the same exposure, even though the energies of the γ rays and the activities of the sources may be quite different. Specifically, the exposure is

$$X = \frac{Q}{m} \tag{6.45}$$

and is measured in the SI units of coulomb per kilogram. More frequently we encounter the *roentgen* unit (R), which is defined as the exposure resulting in an ionization charge of 1 electrostatic unit (the cgs unit of electric charge, in terms of which the electronic charge e is 4.80×10^{-10} electrostatic unit) in 1 cm^3 of air at 0°C and 760 mm pressure (corresponding to a mass of 0.001293 g). Thus

$$1\ \mathrm{R} = \frac{1\ \mathrm{esu}}{0.001293\ \mathrm{g}} = 2.58 \times 10^{-4}\ \mathrm{C/kg}$$

Assigning one unit of electric charge to each ion, an exposure of 1 R means that $(2.58 \times 10^{-4}\ \mathrm{C/kg})/1.60 \times 10^{-19}\ \mathrm{C} = 1.61 \times 10^{15}$ ions are formed per kg of air, or 2.08×10^9 ions per cm^3. It takes on the average about 34 eV to form an ion in air, and thus an exposure of 1 R results in an energy absorption by the air of 7.08×10^{10} eV/cm^3 or 0.113 erg/cm^3, or 88 erg/g.

The ionization produced by a γ ray depends on its energy. With about 34 eV needed to produce each ion in air, a 1-MeV γ ray can be expected to produce, on the average, about 30,000 ions. A radioactive source of a given activity will generally produce many different γ rays with different intensities and energies. The exposure resulting from this source will depend on the number of decays and also on the intensities and energies of each of the γ rays, and the exposure rate (exposure per unit time) will depend on the activity of the source. It will also depend on how far we are from the source; if we imagine that we are to measure the ionization produced in 1 cm^3 of air, that ionization will obviously depend on whether we hold that volume of air very close to the radioactive source or very far away. We can therefore write

$$\frac{\Delta X}{\Delta t} = \Gamma \frac{\mathscr{A}}{d^2} \tag{6.46}$$

where $\Delta X/\Delta t$ is the exposure rate, $\mathscr{A}$ is the activity, d is the distance from the source, and Γ is a constant, the *specific γ-ray constant*, which depends on the details of γ-ray emission of each radionuclide (the fraction of γ rays with each particular energy and the ionizing ability of photons of that particular energy). It is customary to take $d = 1$ m as a standard distance for measuring the relation-

Table 6.3 Specific γ-Ray Constants for Various Radioisotopes[a]

Nuclide	$t_{1/2}$	γ-Ray Energy (MeV) and Abundance (%)	Γ
^{22}Na	2.6 y	0.511 (181), 1.275 (100)	1.20
^{24}Na	15.02 h	1.369 (100), 2.754 (100)	1.84
^{59}Fe	44.6 d	0.143 (1), 0.192 (3), 1.099 (56), 1.292 (44)	0.60
^{57}Co	270 d	0.014 (9), 0.122 (85), 0.136 (11)	0.059
^{60}Co	5.27 y	1.173 (100), 1.333 (100)	1.28
^{131}I	8.06 d	0.08 (2), 0.284 (6), 0.364 (82), 0.637 (7), 0.723 (2)	0.22
^{137}Cs	30.1 y	0.032 (8), 0.662 (85)	0.32
^{198}Au	2.7 d	0.412 (95), 0.676 (1)	0.23
^{226}Ra and daughters			0.84

[a] Units for Γ are R · m^2/h · Ci. Note the relationship between Γ and the energy and intensity of the γ rays.

ship between exposure rate and activity, and thus Γ has units of $(\text{R/h})/(\text{Ci/m}^2)$. Some representative values of Γ are given in Table 6.3.

Materials other than air exposed to ionizing radiation will differ in their rate of energy absorption. It is therefore necessary to have a standard for defining the energy absorption by ionization in different materials. This quantity is called the *absorbed dose D* of the material and measures the energy deposited by ionizing radiation per unit mass of material. The commonly used unit of absorbed dose is the *rad* (*r*adiation *a*bsorbed *d*ose) equal to an energy absorption of 100 ergs per gram of material. (Thus 1 R = 0.88 rad in air.) The SI unit for absorbed dose is the *gray* (Gy), equal to the absorption of 1 joule per kilogram of material, and so 1 Gy = 100 rad.

To define standards for radiation protection of human beings, it is necessary to have some measure of the biological effects of different kinds of radiations. That is, some radiations may deposit their energy over a very long path, so that relatively little energy is deposited over any small interval (say, of the size of a typical human cell); β and γ rays are examples of such radiations. Other types of radiations, α particles for instance, lose energy more rapidly and deposit essentially all of their energy over a very short path length. The probability of cell damage from 1 rad of α radiation is thus far greater than that from 1 rad of γ radiation. To quantify these differences, we define the *relative biological effectiveness* (RBE), as the ratio of the dose of a certain radiation to the dose of X rays *that produces the same biological effect*. Values of the RBE range from 1 to about 20 for α radiation. Since the RBE is a relatively difficult quantity to measure, it is customary to work instead with the *quality factor* (QF), which is calculated for a given type (and energy) of radiation according to the energy deposited per unit path length. Radiations that deposit relatively little energy per unit length (β's and γ's) have QF near 1, while radiations that deposit more energy per unit length (α's) have QF ranging up to about 20. Table 6.4 shows some representative values of QF.

Table 6.4 Quality Factors for Absorbed Radiation

Radiation	QF
X rays, β, γ	1
Low-energy p, n (~ keV)	2–5
Energetic p, n (~ MeV)	5–10
α	20

Table 6.5 Quantities and Units for Measuring Radiation

Quantity	Measure of	Traditional Unit	SI Unit
Activity ($\mathscr{A}$)	Decay rate	curie (Ci)	becquerel (Bq)
Exposure (X)	Ionization in air	roentgen (R)	coulomb per kilogram (C/kg)
Absorbed dose (D)	Energy absorption	rad	gray (Gy)
Dose equivalent (DE)	Biological effectiveness	rem	sievert (Sv)

The effect of a certain radiation on a biological system then depends on the absorbed dose D and on the quality factor QF of the radiation. The *dose equivalent* DE is obtained by multiplying these quantities together:

$$\mathrm{DE} = D \cdot \mathrm{QF} \tag{6.47}$$

The dose equivalent is measured in units of *rem* (*r*oentgen *e*quivalent *m*an) when the dose D is in rads. When the SI unit of gray is used for D, then the dose equivalent is in *sievert* (Sv). Previously we noted that 1 Gy = 100 rad, and so it follows that 1 Sv = 100 rem.

We therefore see that "strength" of radiation has many different ways of being defined, depending on whether we wish to merely count the rate at which the decays occur (activity) or to measure the effect on living systems (dose equivalent). Table 6.5 summarizes these various measures and the traditional and SI units in which they are expressed.

Standards for radiation exposure of the general public and of radiation workers are specified in rems over a certain period of time (usually per calendar quarter and per year). From natural background sources (cosmic rays and naturally occurring radioactive isotopes, such as the uranium and thorium series and ^{40}K) we receive about 0.1–0.2 rem per year. The International Commission on Radiation Protection (ICRP) has recommended limiting annual whole-body absorbed dose to 0.5 rem per year for the general public and 5 rem per year for those who work with radiation. By way of contrast, the dose absorbed by a particularly sensitive area of the body, the bone marrow, is about 0.05 rem for a typical chest X ray and 0.002 rem for dental X rays. Unfortunately, the physiological effects of radiation exposure are difficult to calculate and to measure, and so the guideline must be to keep the exposure as low as possible. (For this reason, many physicians no longer recommend chest X rays as a part of the regular annual physical examination, and dentists often place a lead apron over the sensitive areas of a patient's body while taking X-ray pictures of the mouth.) Although the evidence is not conclusive, there is reason to believe that the risk of radiation-induced cancers and genetic damage remains at even very low doses while other effects, such as cataracts and loss of fertility, may show a definite threshold of exposure below which there is no risk at all. Much of our knowledge in this area comes from studies of the survivors of the nuclear weapons exploded over Hiroshima and Nagasaki in World War II, from which we know that there is virtual certainty of death following a short-term dose of 100 rem, but the evidence regarding the linear relationship between dose and risk is less clear. The effects of long-term, low-level doses are still under active debate, with serious consequences for standards of radiation protection and for the health of the general public.

REFERENCES FOR ADDITIONAL READING

The quantum mechanics of decay processes is treated in more detail in M. G. Bowler, *Nuclear Physics* (Oxford: Pergamon, 1973); see especially Sections 3.1 and 3.2.

A more complete treatment of radioactive decay series can be found in R. D. Evans, *The Atomic Nucleus* (New York: McGraw-Hill, 1955), Chapter 15.

Radioactive dating of the solar system has been reviewed by L. T. Aldrich and G. W. Wetherill, *Ann. Rev. Nucl. Sci.* **8**, 257 (1958), and more recently by G. W. Wetherill, *Ann. Rev. Nucl. Sci.* **25**, 283 (1975).

For more information on radioactivity in the atmosphere and in the oceans, see D. Lal and H. E. Suess, *Ann. Rev. Nucl. Sci.* **18**, 407 (1968).

Additional information on radiation exposure can be found in many references on health physics. See, for example, E. Pochin, *Nuclear Radiation: Risks and Benefits* (Oxford: Clarendon, 1983).

PROBLEMS

1. Three radioactive sources each have activities of 1.0 μCi at $t = 0$. Their half-lives are, respectively, 1.0 s, 1.0 h, and 1.0 d. (a) How many radioactive nuclei are present at $t = 0$ in each source? (b) How many nuclei of each source decay between $t = 0$ and $t = 1$ s? (c) How many decay between $t = 0$ and $t = 1$ h?
2. Naturally occurring samarium includes 15.1% of the radioactive isotope ^{147}Sm, which decays by α emission. One gram of natural Sm gives 89 ± 5 α decays per second. From these data calculate the half-life of the isotope ^{147}Sm and give its uncertainty.
3. Among the radioactive products emitted in the 1986 Chernobyl reactor accident were ^{131}I ($t_{1/2} = 8.0$ d) and ^{137}Cs ($t_{1/2} = 30$ y). There are about five times as many ^{137}Cs atoms as ^{131}I atoms produced in fission. (a) Which isotope contributes the greater activity to the radiation cloud? Assume the reactor had been operating continuously for several days before the radiation was released. (b) How long after the original incident does it take for the two activities to become equal? (c) About 1% of fission events produce ^{131}I, and each fission event releases an energy of about 200 MeV. Given a reactor of the Chernobyl size (1000 MW), calculate the activity in curies of ^{131}I after 24 h of operation.

4. Consider a chain of radioactive decays $1 \to 2 \to 3$, where nuclei of type 3 are stable. (a) Show that Equation 6.31 is the solution to Equation 6.29. (b) Write a *differential equation* for the number of nuclei of type 3 and solve the differential equation for $N_3(t)$. (c) Evaluate $N_1(t) + N_2(t) + N_3(t)$ and interpret. (d) Examine N_1, N_2, and N_3 at small t, keeping only linear terms. Interpret the results. (e) Find the limits of N_1, N_2, and N_3 as $t \to \infty$ and interpret.
5. The human body contains on the average about 18% carbon and 0.2% potassium. Compute the intrinsic activity of the average person from ^{14}C and ^{40}K.
6. A radioactive isotope is prepared by a nuclear reaction in a cyclotron. At the conclusion of the irradiation, which lasts a very short time in comparison with the decay half-life, a chemical procedure is used to extract the radioactive isotope. The chemical procedure takes 1 h to perform and is 100% efficient in recovering the activity. After the chemical separation, the sample is counted for a series of 1-min intervals, with the following results ($t = 0$ is taken to be the conclusion of the irradiation):

t (min)	Decays / min	t (min)	Decays / min	t (min)	Decays / min
62.0	592	112.0	290	163.0	125
68.0	527	120.0	242	170.0	110
73.0	510	125.0	215	175.0	109
85.0	431	130.0	208	180.0	100
90.0	380	138.0	187		
97.0	353	144.0	177		
101.0	318	149.0	158		
105.0	310	156.0	142		

(a) Plot these data on semilog paper and determine the half-life and initial ($t = 0$) activity from your graph. Show the range of uncertainty of each data point, and try to estimate the resulting uncertainty in the half-life. (b) Use an analytic procedure to do a linear least-squares fit of the data (in the form of log N vs t) and determine the half-life and its uncertainty. Formulas for linear least-squares fits can be found in K. S. Krane and L. Schecter, *Am. J. Phys.* **50**, 82 (1982).

7. A sample of a certain element with two naturally occurring isotopes becomes activated by neutron capture. After 1 h in the reactor, it is placed

in a counting room, in which the total number of decays in 1 h is recorded at daily intervals. Here is a summary of the recorded data:

Time (d)	No. Decays	Time (d)	No. Decays
0	102,515	20	2372
1	79,205	40	1421
2	61,903	60	1135
3	48,213	80	862
4	37,431	100	725
5	29,367	120	551
6	23,511	140	462
7	18,495	160	359
8	14,829	180	265
9	11,853	200	225
10	9,595		

From these data, determine the half-lives and initial activities of the two components. What is the element?

8. Consider a simple decay process in which an initial number N_0 of radioactive nuclei of type A decay to stable nuclei of type B. In a time interval from t_1 to $t_1 + \Delta t$, how many decays will occur? Solve this problem in two ways: (1) use Equation 6.10, and (2) use the difference between $N(t_1)$ and $N(t_1 + \Delta t)$. *Note*: Only the first of these methods is correct in general; see the next problem.

9. Consider a decay process $A \rightarrow B \rightarrow C$, in which $N_A(t = 0) = N_0$, and $N_B(t = 0) = N_C(t = 0) = 0$. How many decays of type B nuclei will be observed between t_1 and $t_1 + \Delta t$? (*Hint:* See the previous problem, and explain why method (2) will not work in this case. Figures 6.6 and 6.7 may also provide convincing evidence.)

10. Nuclei of type A, produced at a constant rate R in a nuclear reaction, decay to type B which in turn decay to stable nuclei C. (a) Set up and solve the differential equations for N_A, N_B, and N_C as functions of the time during which the reaction occurs. (b) Evaluate the sum $N_A + N_B + N_C$ and interpret.

11. The radioactive isotope ^{233}Pa ($t_{1/2} = 27.0$ d) can be produced following neutron capture by ^{232}Th. The resulting ^{233}Th decays to ^{233}Pa with a half-life of 22.3 min. Neutron capture in 1 g of ^{232}Th, in the neutron flux from a typical reactor, produces ^{233}Th at a rate of $2.0 \times 10^{11}\ \text{s}^{-1}$. (a) At the end of 1 h of irradiation, what are the resulting activities of ^{233}Th and ^{233}Pa? (b) After 1 h of irradiation, the sample is placed in storage so that the ^{233}Th activity can decay away. What are the ^{233}Th and ^{233}Pa activities after 24 h and after 48 h of storage? (c) The ^{233}Pa decay results in ^{233}U, which is itself radioactive ($t_{1/2} = 1.6 \times 10^5$ y). After the above sample has been stored for 1 year, what is the ^{233}U activity? (*Hint:* It should not be necessary to set up an additional differential equation to find the ^{233}U activity.)

12. An initial activity of nucleus A decays to B, which in turn decays to stable nucleus C. (a) Discuss qualitatively why it must be true at short times that $\mathscr{A}_A > \mathscr{A}_B$, while at long times $\mathscr{A}_B > \mathscr{A}_A$. (b) There must therefore be a time T at which $\mathscr{A}_A = \mathscr{A}_B$. Find T in terms of the decay constants λ_A and λ_B.

13. The decay chain $^{139}\text{Cs} \rightarrow {}^{139}\text{Ba} \rightarrow {}^{139}\text{La}$ is observed from an initially pure sample of 1 mCi of ^{139}Cs. The half-lives are ^{139}Cs, 9.5 min; ^{139}Ba, 82.9 min; ^{139}La, stable. What is the maximum ^{139}Ba activity and when does it occur?

14. In the decay process $^{235}\text{U} \rightarrow {}^{231}\text{Th} \rightarrow {}^{231}\text{Pa}$ ($t_{1/2} = 7.04 \times 10^8$ y for ^{235}U; $t_{1/2} = 25.5$ h for ^{231}Th) plot the ^{235}U and ^{231}Th activities as a function of time from $t = 0$ to $t = 100$ h. Assume the sample initially consists of 1.0 mCi of ^{235}U. Discuss the condition of secular equilibrium in this decay process.

15. The α decay of ^{238}U ($t_{1/2} = 4.47 \times 10^9$ y) leads to 24.1-d ^{234}Th. A sample of uranium ore should reveal ^{234}Th activity in secular equilibrium with the parent. What would be the ^{234}Th activity in each gram of uranium?

16. Prepare a diagram similar to Figure 6.10 showing the $4n + 2$ natural radioactive series.

17. The radioactive decay of ^{232}Th leads eventually to stable ^{208}Pb. A rock is determined to contain 3.65 g of ^{232}Th and 0.75 g of ^{208}Pb. (a) What is the age of the rock, as deduced from the Th/Pb ratio? (b) If the rock is large, the α particles emitted in the decay processes remain trapped. If the rock were pulverized, the α's could be collected as helium gas. At 760 mm and 0°C, what volume of helium gas could be collected from this rock?

18. It is desired to determine the age of a wood timber used to construct an ancient shelter. A sample of the wood is analyzed for its ^{14}C content and gives 2.1 decays per minute. Another sample of the same size from a recently cut tree of the same type gives 5.3 decays per minute. What is the age of the sample?

19. Show that the present ^{14}C content of organic material gives it an activity of about 15 decays per minute per gram of carbon.

20. What is the probability of a ^{14}C decay taking place in the lungs during a single breath? The atmosphere is about 0.03% CO_2, and in an average breath we inhale about 0.5 liter of air and exhale it about 3.5 s later.

21. (a) What is the γ-ray flux (γ's per unit area) a distance of 1.0 m from a 7.5-mCi source of ^{60}Co?

(b) How many ions per minute are produced in a cubic centimeter of air at that distance?